

THE TRAPPING OF NOBLE GASES BY THE IRRADIATION AND WARMING OF INTERSTELLAR

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At the low temperatures typical of interstellar dense molecular clouds ($T = 10\text{--}25\text{ K}$), most molecules will freeze out onto grains where they will be exposed to ionizing radiation in the form of UV photons and cosmic rays. The energetic processing of these ices leads to the production of more complex and refractory organic residues (see [1] for a recent review).

Recently, we have begun working to see if these processes could account for the enigmatic chondritic noble gases found in meteorites. There are several properties of the chondritic noble gases that must be matched by any model that attempts to explain their origin [2]: 1) The primary carrier is carbonaceous. 2) Compared to solar abundances, the gases are elementally fractionated in favor of the heavier gases (i.e., the Kr/Xe ratio is less than the solar ratio). 3) The gases are sited near the surfaces of grains, but are not released (during stepwise heatings) until temperatures of $1000\text{ }^{\circ}\text{C}$ or more. 4) The noble gases are relatively abundant (carbonaceous residues typically contain roughly $3 \times 10^{-7}\text{ cm}^3\text{STP/gm}$ [3]). 5) Despite the elemental fractionation, there is little or no isotopic fractionation compared to the solar wind.

A number of experiments have tried to reproduce the chondritic noble gas pattern, but none has yet succeeded in explaining all of the points above. For example, Wacker *et al.* [4] managed to reproduce the siting and fractionation properties in a carbonaceous carrier, but fell short of the trapping efficiency required to generate the observed abundances, assuming that trapping occurred within the solar nebula. Nichols *et al.* [5] managed to reproduce the abundances needed by using analogs of interstellar silicate "smokes", but the trapping resulted in isotopic fractionation, and was into silicates, not carbon.

We have studied realistic analogs of interstellar ices and the residues they produce when irradiated with UV photons in an attempt to see whether the properties of chondritic noble gases could be reproduced by low temperature interstellar processes. We have analyzed a total of 13 samples (including controls). The samples were made at the Astrochemistry Laboratory at NASA's Ames Research Center using well established techniques [6]. Each sample consists of a piece of high-purity Ni foil, which has been exposed to some combination of ices, noble gases, polycyclic aromatic hydrocarbons (PAHs), and ultraviolet irradiation at low (10 K) temperatures. Typically, a layer of mixed-molecular ice containing $\text{H}_2\text{O}:\text{CH}_3\text{OH}:\text{CO}:\text{NH}_3 = 20:10:2:2$, noble gases $\text{He}:\text{Ne}:\text{Ar}:\text{Kr}:\text{Xe} = 1:1:1:1:1$, and a PAH is laid down, followed by ultraviolet irradiation with a hydrogen lamp, followed by deposition of another ice layer, etc. The $\text{H}_2\text{O}:\text{CH}_3\text{OH}:\text{CO}:\text{NH}_3 = 20:10:2:2$ ice composition was chosen because it provides a good fit to the infrared absorption spectra of many interstellar dense molecular clouds [6]. After the deposition of many layers (typically 10 to 20), the entire sample is warmed. The resulting organic residues that remained at room temperature are sealed in dry nitrogen for shipping to the University of Arizona. Samples are typically exposed to atmosphere for only a few minutes before shipping and during loading into the mass spectrometer extraction line. Samples are heated to $130\text{--}150\text{ }^{\circ}\text{C}$ overnight to expel any adsorbed contamination, and then heated to $1000\text{ }^{\circ}\text{C}$ and $1500\text{ }^{\circ}\text{C}$ and the released gases analyzed in a VG5400 mass spectrometer.

In Table 1, we reported Kr and Xe abundances in the samples analyzed, along with a description of the experimental procedure used to generate each foil. We report only the gas from the $1000\text{ }^{\circ}\text{C}$ extractions, since we have never seen any gas release significantly above background levels at the higher temperature. Also, although we have analyzed He and Ne in some samples, and Ar in almost all, we have not seen any evidence of any of these gases having been trapped. He and Ne would not be expected to be trapped efficiently in our samples since they should not stick efficiently into the original 10 K ice samples. The non-detection of Ar may be due, in part, to its higher backgrounds in the mass spectrometer and due to its low abundance in the original ices relative to Kr and Xe.

In our earliest experiments we attempted to simultaneously produce the sample foils and a number of control foils. Unfortunately, this process led to some confusing results, apparently because of migration of volatiles on the cold head during deposition (samples were mounted on different sides of the same cold head. As a result, we have found it necessary to produce samples and their controls individually. The most recent results (8706-8708) are quite promising: we trapped more Xe than Kr, and the abundance of Xe in 8708 is approaching that in chondritic carbonaceous residues (it is still a factor of 20 too low, but this is still $\sim 10^3$ times higher than the best

previous efforts exploring nebular processes!). Furthermore, the variations in the amounts trapped in those three experiments make sense chemically: The UV irradiation in 8706 corresponded to an interstellar exposure equal to, or greater than, the lifetime of a typical dense molecular cloud and was probably long enough to break down the PAHs in the original ice mixture. In contrast, in 8708 the shorter UV exposures and the addition of butadiene, which should enhance cross-linking, should and did increase the trapping in that sample. Because of the small size of our samples and the small total amounts of gas in them, we have not yet been able to measure isotopic fractionations, if any, in the trapped gases. Some of our earliest experiments suggested that the upper limit on the fractionation is comparable to, or slightly smaller than, that observed by [4]. However, the early experiments apparently suffered from sample 'cross-talk' (see above) and we do not currently consider this issue to be resolved.

Although extremely promising, our results still fall somewhat short of fully explaining chondritic noble gases. In particular, we still haven't trapped quite as much gas as is seen in carbonaceous residues, and the gas we do trap does not seem to be as retentive as the chondritic gases are. In addition, we are still trying to assess the effects of the various stages of sample handling (exposure to air, heat, etc.). Future work will address these issues.

References: [1] S.A. Sandford (1996) *Meteoritics and Planetary Science* **31**, 449-476; [2] T.D. Swindle (1988) In *Meteorites and the Early Solar System*, pp. 535-564; [3] Ott et al. (1981) *Geochim. Cosmochim. Acta* **45**, 1751-1788; [4] J.F. Wacker (1989) *Geochim. Cosmochim. Acta* **53**, 1421-1433; [5] R.H. Nichols Jr. *et al.* (1992) *Meteoritics* **27**, 555-559; [6] M.P. Bernstein *et al.* (1996) *Astrophys. J.* **454**, 327-344.

Table 1 - Noble Gases in Residues Produced by the UV Irradiation of Interstellar Ice Analogs

Expt.	Date Run	Compounds	UV	Layers	Mass	⁸⁴ Kr	¹³² Xe	Kr/Xe
8703	6/96	I,NG,C	3.5	20	<0.60	>0.06	>0.63	0.10
8704	6/96	NG	3.5	10	-	<0.04	<0.04	-
8705	10/96	I,NG,C	3.8	20	0.30	0.05	0.12	0.42
8706	12/96	I,NG,PB	12.0	20	0.20	<0.055	0.23	<0.24
8707	1/97	I,NG,PB	0.75	20	0.28	0.24	1.48	0.16
8708	1/97	I,NG,PB,B	0.75	10	0.05	0.54	16.1	0.034

Compounds: I-Ices (see text); NG-Noble gases; PB-Pyrenebutyric acid; C-Coronene; B-Butadiene
 UV in hours per layer, mass in mg (all masses before 8705 are estimates), gas amounts in
 10⁻⁹ cm³STP/gm except for 8704 which is given in 10⁻¹² cm³STP.